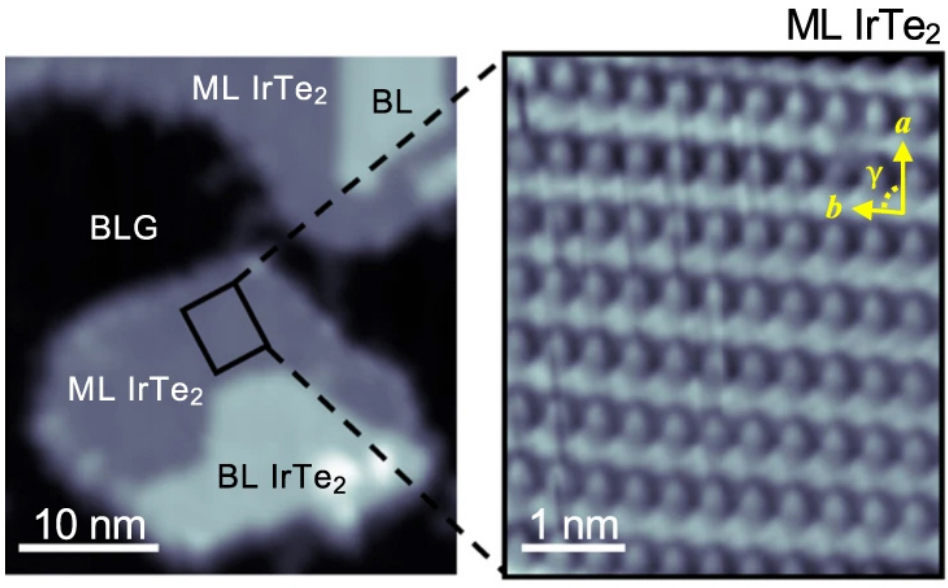
A Novel Insulating State Emerges in a 2D Material



Left: Scanning-tunneling microscope (STM) image of iridium ditelluride ($IrTe_2$), showing both monolayer (ML) and bilayer (BL) regions, on a bilayer graphene (BLG) substrate. Right: Atomically resolved STM image of a section of the monolayer region.

Scientific Achievement

Using the Advanced Light Source (ALS), researchers found a unique insulating state in an atomically thin material, driven by the combined effects of lattice–charge interactions and atomic-bond formation.

Significance and Impact

The work provides a better understanding of charge ordering in two-dimensional materials and opens up new possibilities for achieving designer electronic properties.

A material smorgasbord

Transition-metal dichalcogenides (TMDs) are materials characterized by atomically thin, weakly bonded layers. The basic TMD building block—a monolayer—consists of a course of transition-metal atoms sandwiched between two sheets of chalcogen atoms (sulfur, selenium, or tellurium). For almost a decade, these "2D" materials have been studied extensively for their novel electronic properties and because TMD monolayers can be easily stripped ("exfoliated") from bulk crystals using just adhesive tape. More on point, the periodic table offers 20 to 30 transition-metal options in addition to the three chalcogens—a veritable smorgasbord of combinations to try in search of desirable material properties.

Within the TMD family, iridium ditelluride ($IrTe_2$) is ideally suited for the systematic study of competing factors that can affect a material's electronic properties. For example, bulk $IrTe_2$ exhibits a series of charge-ordered states upon cooling, all while maintaining a metallic nature. Thin films of $IrTe_2$ have recently been shown to exhibit superconductivity as a function of thickness. And the relatively small spacing between $IrTe_2$ layers makes it a good candidate for studying the effects of interlayer coupling.

From bilayer to monolayer

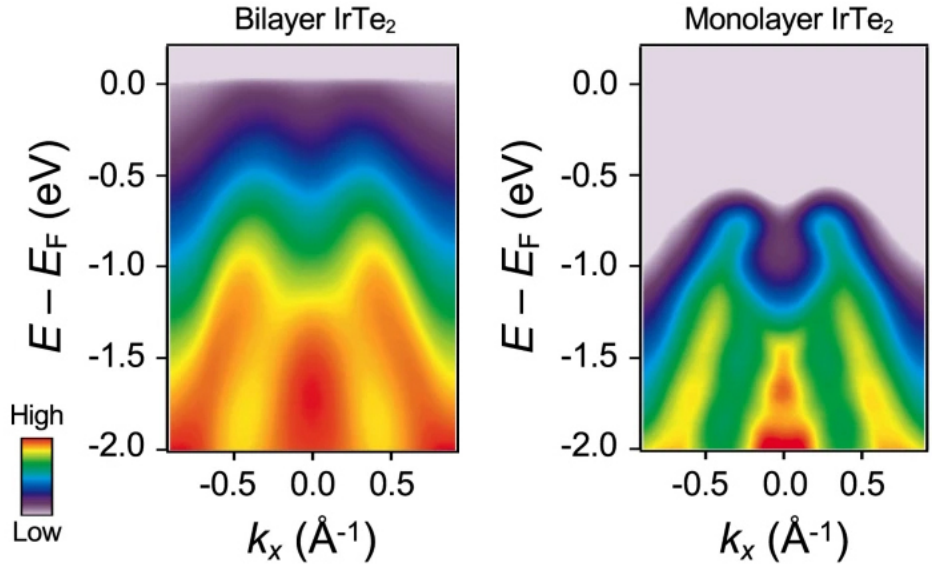
In this work, researchers synthesized bilayer and monolayer $IrTe_2$ samples and characterized their atomic and electronic structures using scanning tunneling microscopy/spectroscopy (STM/STS) at UC Berkeley and angle-resolved photoemission spectroscopy (ARPES) at ALS Beamline 10.0.1. The material synthesis was done using molecular-beam epitaxy (MBE) in a sample-preparation chamber connected to the beamline under ultrahigh vacuum. The system allows for the controlled growth of pristine samples using a variety of materials, yielding clear ARPES data for analysis.

The results showed that monolayer $IrTe_2$ develops a large band gap that's an order of magnitude larger than is typical for TMD systems, transforming the material into an insulator through the removal of a single layer. To better understand this dramatic transition, the researchers performed first-principles calculations to explore various explanations.

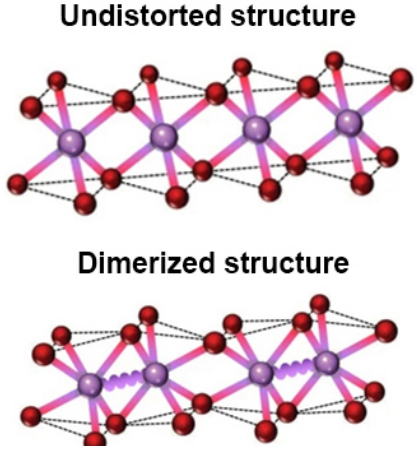
Turbo-charged charge-density waves

Charge-density waves are a type of electronic order in solids: they are modulations in electron density with a periodicity of a few lattice constants. In $IrTe_2$, charge-density waves can also have a reverse effect on the lattice, nudging atoms in particular directions. Theoretical calculations suggested that, in the monolayer, pairs of Ir atoms are pushed and pulled together by a positive feedback loop between a classic charge-density wave and the tendency of the pairs to bond covalently with each other in the absence of valence electrons supplied by an adjacent layer. The researchers concluded that this strong dimer ground state, experimentally supported by both the ARPES and STM data, explains the abrupt appearance of the large band gap in the monolayer's electronic structure.

Overall, the findings provide important insights into the subtle balance of interactions having similar energy scales that occurs in the absence of strong interlayer coupling. They also establish monolayer $IrTe_2$ as a unique large-gap insulator and a useful platform for investigating charge order in layered 2D materials, offering new opportunities for the discovery and control of novel electronic phases.



Left: The ARPES band structure for bilayer $IrTe_2$ clearly exhibits a metallic state, with a band crossing the Fermi energy ($E = E_F$). In contrast, the corresponding ARPES data for monolayer $IrTe_2$ shows an insulating state, with the valence-band maximum at about 0.7 eV below E_F. The gap persists up to 300 K with no change in magnitude.



Depiction of an undistorted $IrTe_2$ structure and a dimerized $IrTe_2$ monolayer. Purple and red balls represent Ir and Te atoms, respectively. Purple wavy lines represent strongly bonded (dimerized) Ir atoms that help drive the material's transition from metal to insulator.

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